Overshooting Effects in the Ordering Dynamics of the Near-Surface Region

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Nonequilibrium thermodynamic properties of first-order phase transformations are of fundamental importance for a detailed understanding of kinetic phenomena in nature and technology. Thus, the growth and coarsening kinetics of (low temperature) bulk ordered phases has been the subject of numerous experimental and theoretical studies.

Within the ordered phase two fundamentally different growth modes exist. The spinodal curve (SP) separates regimes associated with metastable and unstable growth kinetics. Unstable (spinodal) growth starts out with homogeneous nucleation where fluctuations on all length scales are unstable, whereas metastable growth is dominated by heterogeneous nucleation at randomly distributed nucleation sites.

Using surface sensitive x-ray line shape analysis we have studied the time-dependent development of order fluctuations into domains in the subsurface region of $Cu_3Au(001)$ following a rapid temperature quench. The time evolution of the in-plane order does not show any particular surface behavior, especially the time evolution is bulk-like. Unusual overshooting effects in the evolution of the perpendicular order parameter are observed for quenches into the bulk spinodal regime that disappear with a relaxation time τ_R which shows the powerlaw behaviour $\tau_R \sim \lambda^2$ (λ is the length scale of the fluctuation). We argue that these surface relaxation phenomena are driven by a rather general interplay of energy- and entropy-dominated relaxation processes. The origin of the observed behavior is the phase-locking of perpendicular order parameter fluctuations by the surface segregation of Au.